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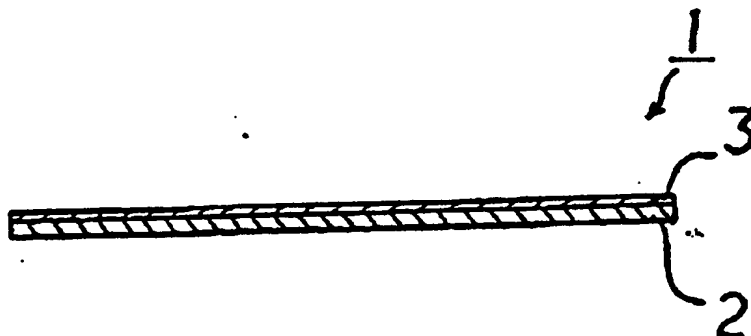
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(54) **Dermal pressure-sensitive adhesive sheet material.**

(57) A dermal pressure-sensitive adhesive sheet material which comprises a backing having provided on one side thereof a pressure-sensitive adhesive layer is disclosed, in which said pressure-sensitive adhesive layer contains a copolymer comprising from 40 to 80% by weight of an alkyl acrylate or methacrylate monomer, from 10 to 50% by weight of an alkoxyl-containing ethylenically unsaturated monomer, and from 1 to 10% by weight of a carboxyl-containing ethylenically unsaturated monomer and having a glass transition point of 250°K or lower and a gel fraction after drying of 25% or more. The dermal pressure-sensitive adhesive sheet material causes no irritation to the skin, behaves like the skin as having low modulus of elasticity, and exhibits high moisture-permeability and excellent resistance to bacteria and water.

FIG 1



EP 0 369 092 A2

DERMAL PRESSURE-SENSITIVE ADHESIVE SHEET MATERIAL**FIELD OF THE INVENTION**

The present invention relates to a dermal pressure-sensitive adhesive sheet material for, for example, adhesive bandage, surgical elements, adhesive plaster, dressing, drape, etc.

BACKGROUND OF THE INVENTION

Conventional dermal pressure-sensitive adhesive sheet materials such as adhesive bandage, comprises a backing having provided on one surface thereof a pressure-sensitive adhesive layer. These sheet materials are used by adhering the same to the skin by means of an adhesive force of the adhesive layer.

The backing in the conventional dermal pressure-sensitive adhesive sheet materials usually comprises polyethylene or polyester. Therefore, these materials do not have a low modulus of elasticity behavior similar to a skin, and cannot follow expansion and contraction of the skin. As a result, a patient feels a cramp on a skin as the patient moves. Thus, use feeling thereof is uncomfortable.

Further, because those materials do not have a sufficient moisture-permeability, sweat is gathered between the skin and the pressure-sensitive adhesive layer to increase the water content of the epidermis more than necessary, which may injure the normal skin. The sweat gathered also tends to reduce adhesive force of the pressure-sensitive adhesive layer to cause slippage or peel of the sheet material off the skin.

In order to overcome the disadvantages attributed to poor moisture-permeability, it is suggested to make the pressure-sensitive adhesive sheet porous or finely porous. However, such porous sheets are not satisfactory from the standpoint of sanitation because fears are entertained that water or bacteria may enter through pores to reach the surface of the skin.

Further, the pressure-sensitive adhesive which contacts a skin of human body generally has a strong irritation to the skin, and long-term contact with the skin induces itching and, in case where the skin is irritable, inflammations or eruptions.

In addition, since the skin has an irregular and complicated surface, it has been very difficult to develop an adhesive having good adhesive properties to the skin combined with satisfactory cohesive force.

Under those circumstances, it has been demanded in the art to develop a dermal pressure-sensitive adhesive sheet material which causes no irritation to the skin, has a low modulus of elasticity behavior similar to the skin, and exhibits high moisture-permeability, excellent bacteria resistance and excellent water resistance.

SUMMARY OF THE INVENTION

Accordingly, an object of the present invention is to provide a dermal pressure-sensitive adhesive sheet material which causes no irritation to the skin, has a low elasticity behavior similar to a skin and exhibits high moisture-permeability, excellent bacteria resistance, and excellent water resistance.

The above object of the present invention can be fully accomplished by a dermal pressure-sensitive adhesive sheet material which comprises a backing having provided on one side thereof a pressure-sensitive adhesive layer, wherein the pressure-sensitive adhesive layer contains a copolymer comprising from 40 to 80% by weight of an alkyl acrylate or methacrylate monomer, from 10 to 50% by weight of an alkoxyl-containing ethylenically unsaturated monomer, and from 1 to 10% by weight of a carboxyl-containing ethylenically unsaturated monomer and having a glass transition point of 250°K or lower and a gel fraction after drying of 25% or more.

BRIEF DESCRIPTION OF THE ACCOMPANYING DRAWING

Figure 1 is a cross section of the dermal pressure-sensitive adhesive sheet material according to the

present invention.

Figure 2 is an illustration of the pressure-sensitive adhesive sheet material of the present invention as applied to the skin.

5

DETAILED DESCRIPTION OF THE INVENTION

Referring to Fig. 1, the dermal pressure-sensitive adhesive sheet material (1) of the present invention comprises a backing (2) having provided on one side thereof a pressure-sensitive adhesive layer (3).

10 The pressure-sensitive adhesive layer contains a copolymer comprising from 40 to 80% by weight of an alkyl acrylate or methacrylate monomer, from 10 to 50% by weight of an alkoxy-containing ethylenically unsaturated monomer, and from 1 to 10% by weight of a carboxyl-containing ethylenically unsaturated monomer.

The reasons for using the alkyl acrylate or methacrylate monomer are that the monomer is a component which imparts adhesive properties to the pressure-sensitive adhesive, there is no irritation to the skin, transparency is excellent, and it is difficult to cause deterioration of adhesive properties even by the use over a long period of time.

In the present invention, the alkyl acrylate or methacrylate must be used in an amount of 40 to 80% by weight based on the weight of the copolymer. The reason is explained below.

20 If the amount of the alkyl acrylate or methacrylate monomer in the copolymer is less than 40% by weight, sufficient adhesive properties cannot be obtained. If it exceeds 80% by weight, a desired cohesive force cannot be obtained.

Specific examples of the alkyl acrylate or methacrylate monomer are butyl acrylate, propyl acrylate, isooctyl acrylate, isononyl acrylate, 2-ethylhexyl acrylate, decyl acrylate and the corresponding methacrylates. These monomers may be used either individually or in combination of two or more thereof.

25 The alkoxy-containing ethylenically unsaturated monomer is a component to impart a moisture-permeability to the adhesive. Further, after drying the copolymer, it undergoes de-alcohol reaction with the carboxyl-containing monomer to form a crosslinked structure.

The alkoxy-containing ethylenically unsaturated monomer must be used in an amount of 10 to 50% by weight. The reason therefor is described below.

If the amount of the alkoxy-containing monomer is less than 10% by weight moisture-permeability and gel fraction is too small to attain a desired cohesive force. If it exceeds 50% by weight, the copolymer becomes too hydrophilic to exhibit sufficient adhesive properties to the skin.

Specific examples of the alkoxy-containing ethylenically unsaturated monomer are methoxy-30 polyethylene glycol, methoxypolyethylene glycol methacrylate, ethoxydiethylene glycol acrylate, butoxydiethylene glycol acrylate, methoxyethyl acrylate, methoxyethyl methacrylate, 3-ethoxypropyl acrylate, ethoxyethyl acrylate, ethoxyethyl methacrylate, butoxyethyl acrylate, and butoxyethyl methacrylate. Those monomers may be used either individually or in combination of two or more thereof.

The carboxyl-containing ethylenically unsaturated monomer functions to improve cohesion of the adhesive and also serves as a reactant in the crosslinking reaction with the alkoxy-containing monomer.

Specific examples of the carboxyl-containing monomer are acrylic acid, methacrylic acid, itaconic acid, and maleic acid.

The carboxyl-containing ethylenically unsaturated monomer must be used in an amount of 1 to 10% by weight. The reason therefor is described below.

45 If the amount of the carboxyl-containing monomer is less than 1% by weight, the cohesive force becomes relatively weak. On the other hand, if it exceeds 10% by weight, not only the cohesive force becomes excessively high, but many polar groups introduced into the copolymer make the adhesive irritative, causing eruptions on the skin when adhered.

In the preparation of the pressure-sensitive adhesive of the present invention, a general polymerization catalyst, such as azobis compounds and peroxide compounds, can be used. The polymerization can be carried out by any technique, such as solution polymerization, emulsion polymerization, and suspension polymerization. For example, a monomer mixture comprising the above-described monomers, a solvent, e.g., ethyl acetate, and a radical generating catalyst are charged in a polymerization vessel, and polymerization is completed in a period of from 10 to 20 hours.

55 If desired, the monomer mixture may further contain a vinyl monomer.

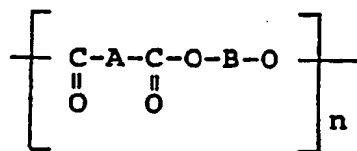
It is required that the resulting copolymer should have a glass transition point of 250° K or lower and a gel fraction after drying of 25% or more. If the glass transition point exceeds 250° K, desired adhesive properties and cohesive force cannot be attained. If the gel fraction after drying is less than 25%, desired

cohesive force cannot be obtained.

Formation of a gel on drying can be accounted for by formation of a crosslinked structure by using the above-described monomers. In general, when a crosslinked structure is formed, adhesive properties are reduced. In the present invention, however, it was confirmed that reduction in adhesive properties is minimized as compared with increase of cohesive force due to the crosslinked structure.

Thickness of the pressure-sensitive adhesive layer is not particularly limited but preferably from 10 to 50 μm . A thickness less than 10 μm tends to have insufficient adhesive properties to the skin, while that exceeding 50 μm tends to have insufficient moisture-permeability. Anyway, a suitable thickness can be determined according to the portion of skin to which the adhesive layer is adhered.

Materials of the backing which can be used in the present invention include polyether urethane, polyester urethane, polyether-polyamide block copolymers, and polyacrylate. Preferred materials are polyether-polyamide block copolymers represented by formula:



wherein A represents a polyamide component selected from nylon 6, nylon 6.6, nylon 10, nylon 11, and nylon 12; B represents a polyether component selected from polyethylene glycol, polypropylene glycol, and polytetramethylene glycol; and n represents a positive integer.

The polyamide component as represented by A can be obtained by polycondensation between a dicarboxylic acid (e.g., terephthalic acid, oxalic acid, adipic acid, sebacic acid, 1-cyclohexydicarboxylic acid) and a diamine (e.g., ethylenediamine, pentamethylenediamine, hexamethylenediamine), polymerization of cyclic lactams (e.g., caprolactam, caprylolactam, caprinolactam, laurinlactam), polycondensation of aminocarboxylic acids, or copolymerization of the above-described cyclic lactams, dicarboxylic acids, and diamines.

The polyether component as represented by B includes polyoxyalkylene glycols, e.g., polyoxyethylene glycol, polyoxypropylene glycol, and polyoxytetramethylene glycol.

A commercially available product (e.g., "PEBAX" a trade name of Toray Industries, Inc.) may be utilized as the above-described block copolymer.

Thickness of the backing is not particularly limited, but preferably ranges from 20 to 100 μm . Too a thin backing is inconvenient on handling such as adhering or peeling. Too a thick backing may lose its softness and interfere with moisture-permeation. Anyway, a proper thickness of the backing can be determined according to the portion of a skin to which the backing is adhered.

When the adhesive sheet material of the present invention is applied to parts having relatively large expansion and contraction of a skin, such as joints, it is preferred that the backing has a tensile strength of from 100 to 500 kg/cm^2 and a 100% modulus of from 10 to 100 kg/cm^2 . If the tensile strength exceeds 500 kg/cm^2 or the 100% modulus exceeds 100 kg/cm^2 , it is difficult to impart a softness to the backing, and when it is adhered on a skin as a dermal pressure-sensitive adhesive sheet material, the sheet material cannot follow the expansion and contraction of a skin, thereby giving physical irritation to the skin to feel uncomfortable feeling, or causing slippage or peel of the sheet material. On the other hand, if the tensile strength is less than 100 kg/cm^2 or 100% modulus is less than 10 kg/cm^2 , the base sheet is too soft to handle. Thus, problems on use occur.

In the case of making the block polymer represented by the above-described formula have the tensile strength and modulus in the above-described ranges, it can be achieved by changing the ratio of a hard segment and a soft segment in the block polymer. That is, it is preferred that the weight percentage of the polyether component B is 50 to 90% by weight based on the total weight of the polyamide component A which is the hard segment component and the polyether component B which is the soft segment component.

However, in the event that the dermal pressure-sensitive adhesive sheet material of the present invention is applied to the skin of parts making relatively small movements, such as thighs, legs, arms, chest, etc., the tensile strength and 100% modulus of the backing are not particularly restricted and can be appropriately selected.

When the dermal pressure-sensitive adhesive sheet material of the present invention is applied to parts

which are easily wet with sweat, such as the palm of a hand and the sole of a foot, it is preferred that the adhesive layer has a moisture-permeability of $1,000 \text{ g/m}^2 \cdot 24 \text{ hrs} \cdot 40^\circ \text{C}$ or more and that the laminate of the backing and the adhesive layer as a whole has a moisture-permeability of $800 \text{ g/m}^2 \cdot 24 \text{ hrs} \cdot 40^\circ \text{C}$ or more.

5 If the moisture-permeability of the pressure-sensitive adhesive layer is less than $1,000 \text{ g/m}^2 \cdot 24 \text{ hrs} \cdot 40^\circ \text{C}$ or the moisture-permeability as a whole of the laminate of backing and pressure-sensitive adhesive layer is less than $800 \text{ g/m}^2 \cdot 24 \text{ hrs} \cdot 40^\circ \text{C}$, there is a tendency that sweat is gathered between the pressure-sensitive adhesive layer and the skin surface to increase the water content of the epidermis more than necessary, which may cause injury of the normal skin, or to reduce the adhesive force of the pressure-sensitive adhesive to cause slippage or peel.

10 In the case that the dermal pressure-sensitive adhesive sheet material is applied to parts having a relatively small sweating, the moisture-permeability of the pressure-sensitive adhesive layer and the dermal pressure-sensitive adhesive sheet material as a whole is not limited to the above-described range and is appropriately selected.

15 The pressure-sensitive sheet according to the present invention can be prepared by directly coating the pressure-sensitive adhesive on a backing, drying the adhesive, and applying a release sheet on the adhesive layer; or coating the pressure-sensitive adhesive on a release sheet, drying the adhesive, and adhering a backing to the adhesive layer. By this procedure, a dermal pressure-sensitive adhesive sheet with the pressure-sensitive adhesive layer protected with the release sheet can be obtained.

20 The structure of the dermal pressure-sensitive adhesive sheet material according to the present invention is as described above. In using the dermal pressure-sensitive adhesive sheet material, as shown in Fig. 2, the adhesive sheet material of the present invention can be adhered to the skin of the affected part utilizing the adhesive force of the pressure-sensitive adhesive layer. The dermal pressure-sensitive adhesive sheet thus adhered can easily follow the movement of the skin, while removing sweat as moisture

25 (4). The dermal pressure-sensitive adhesive sheet material according to the present invention exhibits the following effects.

The pressure-sensitive adhesive layer contains a copolymer comprising 40 to 80% by weight of an alkyl acrylate or methacrylate monomer, 10 to 50% by weight of an alkoxy-containing ethylenically unsaturated monomer and 1 to 10% by weight of a carboxyl-containing ethylenically unsaturated monomer. Therefore, the adhesive is non-irritative to the skin and excellent in transparency, and the adhesion thereof is not reduced during long-term use. Further, since the copolymer has a glass transition point of 250°K or less and a gel fraction after drying of 25% or more, satisfactory adhesive property and cohesive force can be obtained, which assure excellent holding properties particularly when the adhesive sheet material is used as an element for fixing medical equipment. Thus, the adhesive sheet material causes no strong irritation to the skin or suffers from no reduction in adhesion during long-term use and are, therefore, excellent in safety and stability.

30 In particular, when the moisture-permeability of the pressure-sensitive adhesive layer is $1,000 \text{ g/m}^2$ or more and that of the laminate of the backing and the adhesive layer is 800 g/m^2 or more, the sweat from the skin is effectively vaporized without being collected between the skin and the pressure-sensitive adhesive layer so that there is no fear that the water content of the epidermis excessively increases to injure a normal skin or the adhesion of the adhesive layer is reduced to cause slippage or peel.

35 Since no pores for moisture escape are required, water or bacteria are not allowed to reach the skin. This also means that the exudate from the affected part would not ooze of the backing. Namely, the adhesive sheet is very sanitary.

40 When, in particular, the backing has a tensile strength of from 100 to 500 kg/cm^2 and a 100% modulus of from 10 to 100 kg/cm^2 , the similarity to the skin in elasticity is further ensured, while retaining easiness on handling such as adhering or peeling, and the backing can further securely follow movement of the skin.

50 The present invention is now illustrated in greater detail by reference to Examples and Comparative Examples, but it should be understood that the present invention is not deemed to be limited thereto. In these examples, all the parts are by weight unless otherwise indicated.

The copolymers and pressure-adhesive sheets prepared in Examples and Comparative Examples were evaluated for physical properties or performance properties according to the following test methods.

55

1) Adhesive Strength of Adhesive Sheet Material:

A 20 mm wide dermal pressure-sensitive adhesive sheet material was subjected to peel test at a peel angle of 180° using a Bakelite plate as an adherend according to JIS Z-1528 to measure an adhesive strength (g/20 mm).

2) Holding Time of Adhesive Sheet:

A dermal pressure-sensitive adhesive sheet material having a size of 10 mm x 20 mm was adhered onto a Bakelite plate. A load of 500 g was applied thereon at 40° C, and the time (minute) until the adhesive sheet fell off the plate was measured. The adhesive sheet material under test was backed with a polyester film to prevent stretching of the sheet material during the load application.

3) Moisture-Permeability of Adhesive Sheet Material:

10 ml of distilled water was put in a glass-made container having an inner diameter of 38 mm and a height of 40 mm, and an adhesive sheet material having a diameter of 50 mm was placed thereon and fixed with its adhesive layer facing downward to cover the opening in. The container was allowed to stand in an air-conditioned room at 40° C and 30% RH for 24 hours, and the loss of water in the container was measured to calculate moisture permeability ($\text{g/m}^2 \cdot 24 \text{ hrs} \cdot 40^\circ \text{C}$).

4) Gel Fraction of Copolymer:

A predetermined amount of a dried polymer was extracted with ethyl acetate for 24 hours, and the residue was weighed. A gel fraction was calculated from formula:

$$\frac{(\text{Weight of Extraction Residue})}{(\text{Weight of Polymer Before Extraction})} \times 100 (\%)$$

EXAMPLE 1

2-Ethylhexyl acrylate	70 parts
Ethoxyethyl acrylate	25 parts
Acrylic acid	5 parts
Ethyl acetate	150 parts
Azoisobutyronitrile	0.3 part

The above components were charged in a polymerization vessel, and the mixture was stirred while displacing the atmosphere with nitrogen. Thereafter, polymerization was conducted for about 10 hours while keeping the inner temperature at 55 to 65° C. The inner temperature was raised up to 70° C, at which the stirring was continued for an additional about 2 hours. The resulting copolymer had a glass transition point (T_g) of 212° K and a gel fraction of 31.5%.

The copolymer was coated on release paper having been treated with silicone to a dry thickness of 25 μm and dried at 130° C for 5 minutes to form a pressure-sensitive adhesive layer. A 35 μm thick polyether-polyamide block copolymer sheet ("PEBAX 3533" a trade name of Toray Industries, Ltd.), one side of which had been subjected to corona discharge treatment, was adhered onto the adhesive layer under pressure to obtain a dermal pressure-sensitive adhesive sheet material.

The adhesive strength, holding time, and moisture-permeability of the resulting adhesive sheet material are shown in Table 1.

EXAMPLE 2

5 A pressure-sensitive adhesive sheet material was prepared in the same manner as in Example 1, except for using the following monomer mixture. The Tg and gel content of the copolymer and the adhesive strength, holding time, and moisture-permeability of the adhesive sheet material are shown in Table 1.

Isononyl acrylate	62 parts
Methoxyethyl acrylate	35 parts
Acrylic acid	3 parts

EXAMPLE 3

15 A pressure-sensitive adhesive sheet material was prepared in the same manner as in Example 1, except for using the following monomer mixture. The Tg and gel fraction of the copolymer and the adhesive strength, holding time, and moisture-permeability of the adhesive sheet material are shown in Table 1.

2-Ethylhexyl acrylate	65 parts
Butoxyethyl acrylate	30 parts
Methacrylic acid	5 parts

COMPARATIVE EXAMPLE 1

25 A pressure-sensitive adhesive sheet was prepared in the same manner as in Example 1, except for replacing ethyl acetate with a mixed solvent of toluene and laurylcaptane (100/0.025 by weight). The Tg and gel fraction of the copolymer and the adhesive strength, holding time, and moisture-permeability of the adhesive sheet material are shown in Table 1.

COMPARATIVE EXAMPLE 2

30 A pressure-sensitive adhesive sheet material was prepared in the same manner as in Example 1, except for changing the drying condition to 95° C and 5 minutes. The Tg and gel fraction of the copolymer and the adhesive strength, holding time, and moisture-permeability of the adhesive sheet material are shown in Table 1.

COMPARATIVE EXAMPLE 3

35 A pressure-sensitive adhesive sheet material was prepared in the same manner as in Example 1, except for using the following monomer mixture. The Tg and gel fraction of the copolymer and the adhesive strength, holding time, and moisture-permeability of the adhesive sheet material are shown in Table 1.

2-Ethylhexyl acrylate	95 parts
Acrylic acid	5 parts

TABLE 1

Example No.	Adhesive Strength	Holding Time	Moisture Permeability	Tg	Gel Fraction
	(g/19mm)		(g/m ² 24 hr 40 °C)	(°C)	(%)
Example 1	420	100 min. or more	950	212	31.5
" 2	500	"	1100	207	28.0
" 3	450	"	880	214	30.5
Comparative Example 1	490	fall in 30 min. (cohesive failure)	1050	212	0
" 2	400	fall in 15 min. (cohesive failure)	950	212	5
" 3	500	fall in 30 min. (cohesive failure)	530	208	0

While the invention has been described in detail and with reference to specific embodiments thereof, it will be apparent to one skilled in the art that various changes and modifications can be made therein without departing from the spirit and scope thereof.

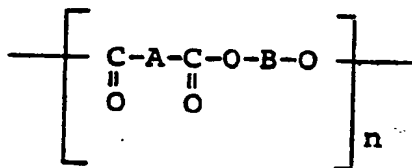
Claims

1. A dermal pressure-sensitive adhesive sheet material which comprises a backing having provided on one side thereof a pressure-sensitive adhesive layer, wherein said pressure-sensitive adhesive layer contains a copolymer comprising from 40 to 80% by weight of an alkyl acrylate or methacrylate monomer, from 10 to 50% by weight of an alkoxy-containing ethylenically unsaturated monomer, and from 1 to 10% by weight of a carboxyl-containing ethylenically unsaturated monomer and having a glass transition point of 250° K or lower and a gel fraction after drying of 25% or more.

2. A dermal pressure-sensitive adhesive sheet material as claimed in claim 1, wherein said pressure-sensitive adhesive layer has a moisture-permeability of 1,000 g/m² or more and said adhesive sheet material has a moisture-permeability of 800 g/m² or more.

3. A dermal pressure-sensitive adhesive sheet material as claimed in claim 1, wherein said backing has a tensile strength of from 100 to 500 kg/cm² and a 100% modulus of from 10 to 100 kg/cm².

4. A dermal pressure-sensitive adhesive sheet material as claimed in claim 1, wherein said backing comprises a polyether-polyamide block copolymer represented by formula:



wherein A represents a polyamide component selected from the group consisting of nylon 6, nylon 6.6, nylon 10, nylon 11, and nylon 12; B represents a polyether component selected from the group consisting of polyethylene glycol, polypropylene glycol, and polytetramethylene glycol; and n represents a positive integer.

FIG 1

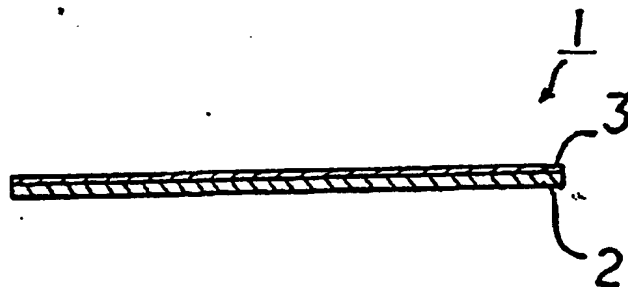
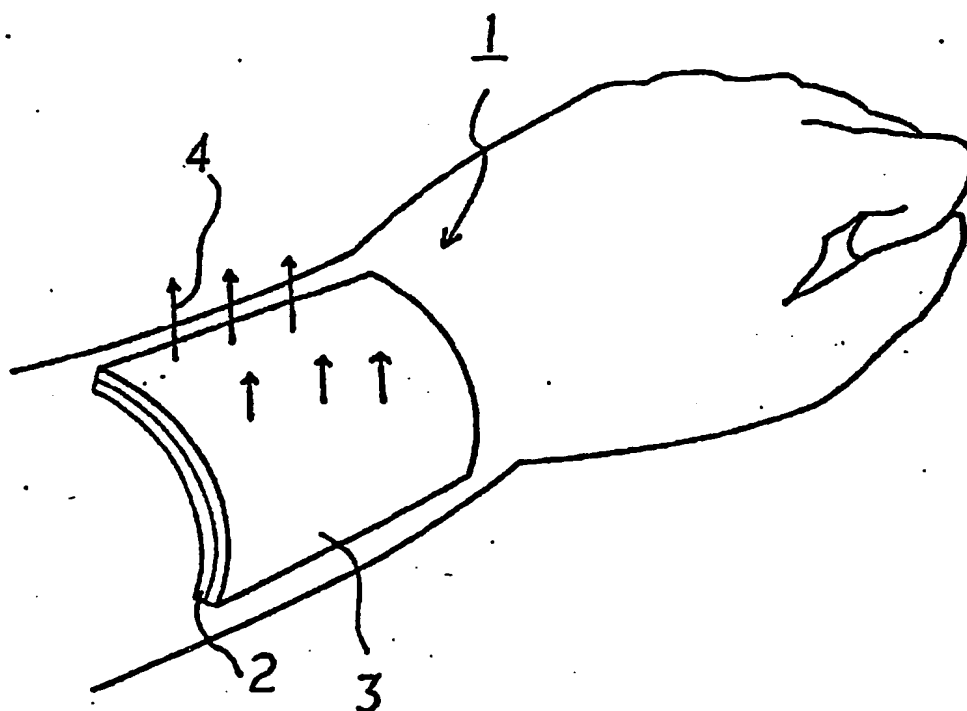
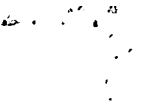


FIG 2





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05.09.90 Bulletin 90/36(71) Applicant: **NITTO DENKO CORPORATION**
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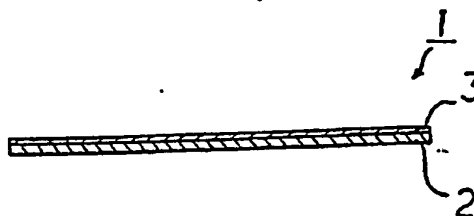
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(54) **Dermal pressure-sensitive adhesive sheet material.**

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FIG 1



EP 0 369 092 A3



European Patent
Office

EUROPEAN SEARCH REPORT

Application Number

EP 89 10 2340

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl. 5)
X	US-A-4 608 249 (S. OTSUKA) * Claims; column 2, line 11 - column 3, line 18; example 3 * ---	1	A 61 L 15/58 C 09 J 133/06
Y	EP-A-0 107 915 (SMITH AND NEPHEW) * Page 11, lines 6-16; claims * ---	3-4	
Y	GB-A-1 473 972 (ATO CHEMIE) * Claims; page 2, lines 10-47 * ---	3-4	
A	EP-A-0 123 465 (SMITH AND NEPHEW) * Claims; example 11 * ---	1-4	
A	WO-A-8 801 877 (SMITH AND NEPHEW) * Example 7; page 7, paragraph 3 - page 8, paragraph 2; page 12, paragraph 3 * ---	2-4	
A	EP-A-0 174 803 (SMITH AND NEPHEW) -----		
			TECHNICAL FIELDS SEARCHED (Int. Cl. 5)
			A 61 L C 09 J
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 19-06-1990	Examiner COUSINS-VAN STEEN G.I.L.
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X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	

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